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Solubility-Limited Concentrations and Aqueous Speciation of U, Pu, Np, Am and Tc: Comparison Between Results of Bruno and Sellin (1992) and Calculations Using GEMBOCHS (version R16)

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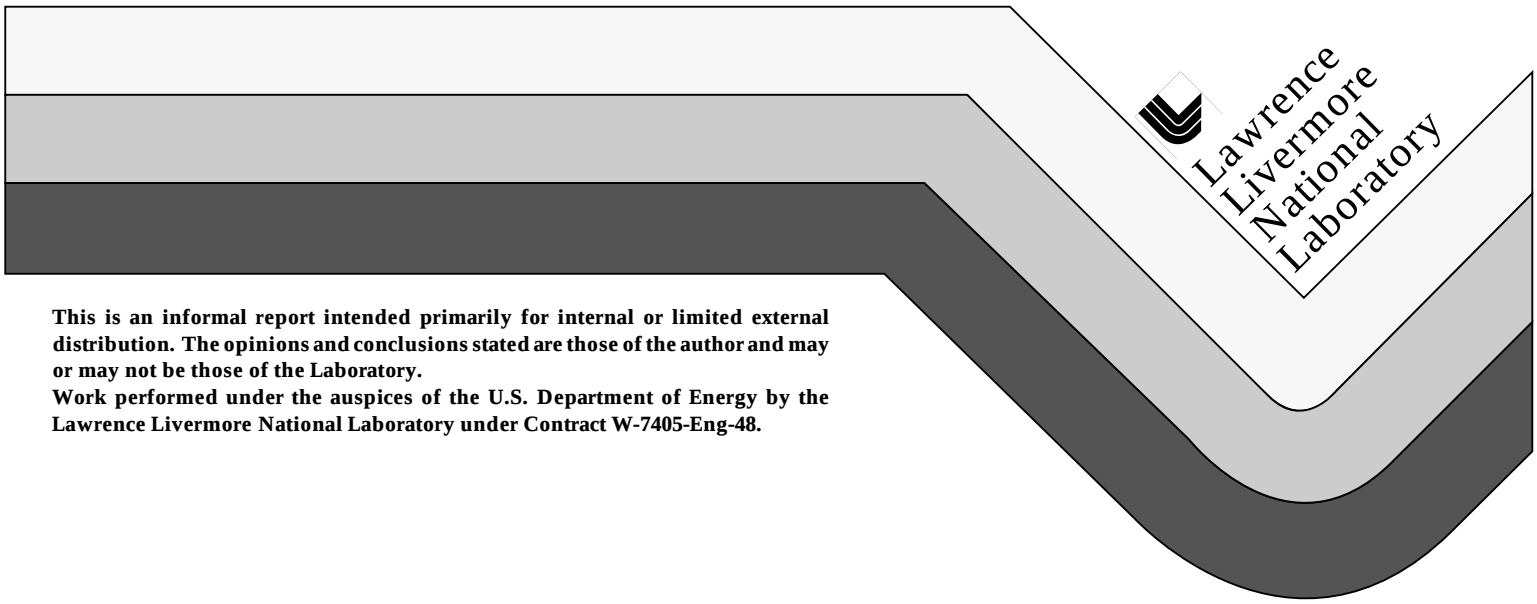

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Solubility-limited concentrations and aqueous speciation of U, Pu, Np, Am and Tc: Comparison between results of Bruno and Sellin (1992) and calculations using GEMBOCHS (version R16)

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Abstract

The aqueous speciation and solubility-limited concentrations of U, Pu, Np, Am and Tc were calculated with EQ3/6 and version comR16 of the GEMBOCHS data base (Wolery, 1992) for comparison to similar calculations made by Bruno and Sellin (1992) for the SKB 91 exercise. Bruno and Sellin utilized data from the older 0288 version of the EQ3/6 data base but substituted their own data sets for U and Pu. Equilibria were computed in representative fresh and saline Finnsjön-waters under oxidizing and reducing conditions.

Comparisons between the two sets of calculations showed that slight discrepancies exist for U because Bruno and Sellin used thermodynamic data from sources that pre-date the Nuclear Energy Agency (NEA) data base (Grenthe et al., 1992). This NEA data base is incorporated into GEMBOCHS. Discrepancy also exists for Pu under reducing conditions because of the choice of thermodynamic data for solid $Pu(OH)_4$. GEMBOCHS predicts Pu concentrations in solution that are about 1 to 2 orders of magnitude greater than Bruno and Sellin's values.

The concentration of Np in the oxidizing saline water computed with GEMBOCHS is 20 times higher than Bruno and Sellin's value. Under reducing conditions, however, GEMBOCHS predicts an order of magnitude less Np in solution. GEMBOCHS computes Am concentrations in solution about 2-3 times larger than those of Bruno and Sellin. Bruno and Sellin's data base lacks the aqueous species $Am(CO_3)_2^-$, although this species occurs only sparingly (< 10 mol%) in the fresh waters.

The overall Tc data bases differ significantly because more recent critical compilations of Tc data have been incorporated into GEMBOCHS since Bruno and Sellin's 0288 version. Nonetheless, results for Tc are broadly similar, although GEMBOCHS predicts Tc concentrations about 1.7 times higher than Bruno and Sellin's values for the reducing waters.

Introduction

Accurate and comprehensive thermodynamic data are required by geochemical modeling codes to simulate the behavior of radionuclides in nuclear waste repositories. The purpose of this report is to compare aqueous speciation and solubility-limited concentrations of U, Pu, Np, Am and Tc calculated by Bruno and Sellin (1992) as part of the SKB 91 exercise with those calculated using EQ3/6 and version comR16 of the GEMBOCHS data base

(Wolery, 1992). Version comR16 contains the latest sets of radionuclide thermodynamic data used by the U.S. Dept. of Energy Yucca Mountain Project as part of the EQ3/6 geochemical modeling code package.

Bruno and Sellin (1992) published tables of the concentrations and aqueous speciation of radionuclides in equilibrium with specified solid phases for four different granitic groundwaters. They used version 0288 of the EQ3/6 computer code package (alternately described as version R54 of the data base), but substituted alternate thermodynamic data for U and Pu from Puigdomenech and Bruno (1988; 1991). EQ3/6 was used in conjunction with version comR16 of the GEMBOCHS thermodynamic data base (hereafter referred to as GEMBOCHS) to try to duplicate Bruno and Sellin's calculations and identify calculational discrepancies that may result from the use of alternate data bases.

Fluid chemistry

Bruno and Sellin (1992) used typical fresh and saline Finnsjön-waters under oxidizing and reducing conditions for fluid chemistry in the SKB 91 exercise. The fresh and saline Finnsjön-waters given by Bruno and Sellin are out of charge balance by about 6-9% and 16% of the total charge, respectively. The waters were deficient in anions relative to cations. No attempt was made to charge balance the waters because the main purpose of this calculation is to compare results with those of Bruno and Sellin, who apparently did not charge balance the waters before their solubility calculations were made. The source of the charge discrepancy should be investigated. The presence of additional carbonate complexes would change computed radionuclide concentrations slightly.

The oxidation state used for the oxidizing waters by Bruno and Sellin is somewhat ambiguous. In the caption to Table 3, they state they use 0.2 atm fugacity of $O_{2(g)}$ (log $f(O_{2(g)}$ = -0.7), whereas in Table 2, they list 650 mV. These expressions of redox state are not equivalent. At 25°C in both the fresh and saline waters with pH values of 6.9 and 7, 650 mV corresponds to log fugacity ($O_{2(g)}$) less than -11. The solubility calculations in this paper were made assuming Eh values of 650 and -200 mV.

Calculational procedure

The total concentration of a radionuclide in solution and its aqueous speciation were calculated by requiring EQ3 to equilibrate the solution with a solid phase containing that element. EQ3 then computed the required total concentration of radionuclide in solution, taking explicit consideration of the aqueous speciation of the radionuclide. All calculations were made at 25°C and 1 bar. The choice of solubility-limiting solids are generally those chosen by Bruno and Sellin (1992). Solids that were supersaturated given the assumed identity of the solubility-limiting solid are noted in the footnotes to Tables 1 and 2. Aqueous species from GEMBOCHS that were suppressed in the simulations are also noted in the table footnotes.

The computed aqueous speciation is dependent on the identity of the solubility-limited solid. For example, the species $UO_2(CO_3)_2^{2-}$ and $UO_2(CO_3)_3^{4-}$ are dominant in the Finnsjön-waters at trace concentrations of U, but the species $(UO_2)_2CO_3(OH)_3^{1-}$ dominates when U concentrations are large, as controlled by schoepite equilibrium. In another example, $TcO(OH)_2^{o}$ is the dominant aqueous species of Tc at trace concentrations of Tc

in solution. However, when equilibrium with $TcO_2 \cdot 2H_2O$(am) is fixed, the species $(TcO(OH)_2)_2^o$ forms as well because of the relatively high solubility of $TcO_2 \cdot 2H_2O$(am). Thus, a different choice of the solubility-limiting phase may change the aqueous speciation if a significantly different concentration of the radionuclide results.

Tables 1 and 2 summarize results for each radionuclide in saline and fresh Finnsjön-waters, respectively, under oxidizing and reducing conditions. Results for each radionuclide are discussed below. For the sake of brevity, calculations with EQ3/6 and GEMBOCHS version comR16 are sometimes referred to as GEMBOCHS.

Uranium

Thermodynamic data

Bruno and Sellin (1992) use the SKBU 1 thermodynamic data base for U to calculate radionuclide solubilities for SKB 91. The data are discussed in Puigdomenech and Bruno (1988). It is sourced mainly from Lemire and Tremaine (1980) and Lemire (1988), with modifications by Puigdomenech and Bruno (1988). Bruno and Sellin state that the data are in agreement with the recommendations of a pre-publication draft of uranium data from the Nuclear Energy Agency (Grenthe et al., 1992).

Thermodynamic data at 25°C for most aqueous species and solids of U in the latest EQ3/6 data bases (GEMBOCHS; versions R16 and later) are sourced from the Nuclear Energy Agency (NEA) data base (Grenthe et al., 1992). The NEA data base has no data for U-silicates except for coffinite. However, U-silicates such as uranophane are common in nature. Thermodynamic data for U-silicates except for coffinite in GEMBOCHS are estimated (Hemingway, 1982; Langmuir, 1978). No experimental determinations existed until the work of Nguyen et al. (1992) who determined Gibbs free energy of formation for soddyite, uranophane, sodium boltwoodite and sodium weeksite. However, this data has not yet been evaluated for inclusion into the comR16 data base. The estimated data are included in GEMBOCHS because U-silicates are potential precipitates in systems containing Si.

The estimated data for haiweeite and soddyite in comR16 are from Hemingway (1982). The experiments of Nguyen et al. (1992) indicate that soddyite is much less stable than the estimated values would suggest. The uranophane value in comR16 was estimated by Langmuir (1978) using phase relationships including (uranophane + CO_2(g) = calcite + SiO_2(aq) + schoepite + water). However, Langmuir used thermodynamic data for schoepite different from that in the NEA data base. I revised the data for uranophane in GEMBOCHS comR16 to be consistent with the NEA data for schoepite, which involved decreasing its free energy of formation by about 1.8 kcal/mol. This change had no effect on the calculational results described in this paper, however, because schoepite was used as the solubility-limiting phase.

The NEA assigns a limiting value of ΔG_f to $U(OH)_5^-$ that makes it stable relative to $U(OH)_4$(aq) above pH=12 (p. 123, Grenthe et al., 1992). $U(OH)_5^-$ was not included in GEMBOCHS because it had a limiting ΔG_f value. Although one must consider the possibility of $U(OH)_5^-$ formation when pH exceeds 12, the pH values in this exercise are far below that value so this species should not be of concern.

The ΔG_f of $UO_2(OH)_2$(aq) is given as $\geq$ -1368 kJ/mol in the NEA data base; its ΔG_f is tabulated in GEMBOCHS as -1368 kJ/mol. The stability of $UO_2(OH)_2$(aq) may thus be overestimated to an unknown extent when using this data. Puigdomenech and Bruno (1988) list ΔG_f for this species as -1359 kJ/mol. This complex was predicted to form in the GEMBOCHS calculations, but not in those by Bruno and Sellin.

Comparison of modeling results

Table 1 compares solubility-limited concentrations of U in fresh and saline Finnsjöwater at 25°C calculated using the LLNL data base GEMBOCHS version comR16, with concentrations from Bruno and Sellin (1992) using the SKB data base, as described in Puigdomenech and Bruno (1988). The solubility-limiting phases, schoepite and UO_2(fuel), yield conservative estimates of radionuclides in solution as a number of more insoluble solid phases are supersaturated under these conditions. Data for UO_2(fuel) is not included in the EQ3/6 data base, but is listed in Puigdomenech and Bruno (1988) and was used in the Bruno and Sellin exercise. Consequently, the data for UO_2(fuel) was temporarily added to the comR16 data base for this exercise.

EQ3 with comR16 predicts higher U concentrations in solution than Bruno and Sellin's calculations for the saline water. Under oxidizing conditions, GEMBOCHS predicts a ten-fold higher U concentration. Part of this difference reflects the inclusion of the aqueous species $(UO_2)_2CO_3(OH)_3^-$ in GEMBOCHS, which is computed to be the dominant aqueous complex of U. This species is not present in Puigdomenech and Bruno (1988) data base used by Bruno and Sellin (1992). Even when this species is suppressed in GEMBOCHS, however, the computed concentration of U equals 1.4×10^{-5} molal, as compared to Bruno and Sellin's 3×10^{-6} molal.

Discrepancies in computed U concentration also exist under reducing conditions for both the fresh and saline waters when both codes predict $U(OH)_4^o$ as the dominant aqueous species. GEMBOCHS predicts a 1.6 fold higher U concentration. The ΔG_f for $U(OH)_4^o$ in GEMBOCHS and Grenthe et al. (1992) is 1 kcal/mol more negative than the value used by Bruno and Sellin, which would account for at least part of the discrepancy.

U concentrations under oxidizing conditions are higher in the fresh waters because they contain greater quantities of HCO_3^- than the saline waters (220 vs. 48 ppm). Computed concentrations are similar under oxidizing conditions in the fresh water, but not in the saline waters.

The speciation of U in the fresh waters changes as U in solution increases from trace amounts, when $UO_2(CO_3)_2^-$ and $UO_2(CO_3)_3^{4-}$ are dominant, to 62 mg/kg in equilibrium with schoepite, when $(UO_2)_2CO_3(OH)_3^-$ sequesters half the U in solution. This aqueous species forms because of the large amounts of U in solution required to force schoepite to equilibrate. Equilibrium with schoepite and UO_2(fuel) yield conservative values; that is, values that will probably not be exceeded for use in bounding performance assessment calculations.

Plutonium

Thermodynamic data

Puigdomenech and Bruno (1991) describe the thermodynamic data used by Bruno and Sellin (1992) in their SKB 91 work. A species-by-species comparison with the current GEMBOCHS data base was not made. Instead, the calculational results were compared to determine the quantitative impact of the differences in the data bases. The differences were then related back to differences in the data bases whenever possible.

Comparison of modeling results

GEMBOCHS predicted Pu concentrations about 1 to 2 orders of magnitude greater than those published by Bruno and Sellin in all four waters. The major part of this discrepancy arises because of the difference in data for the solid $Pu(OH)_4$. Both GEMBOCHS and Puigdomenech and Bruno (1991) claim Lemire and Tremaine (1980) as their data source. The data in GEMBOCHS match Lemire and Tremaine's published data. However, Puigdomenech and Bruno list the free energy of formation from the elements of $Pu(OH)_4$(am) as -342.97 kcal/mol, versus -340.82 for the solid $Pu(OH)_4$ in Lemire and Tremaine and GEMBOCHS.

A difference of 2.15 kcal/mol translates to a change in log K of 1.58 at 25°C for any reaction involving 1 mole of the solid $Pu(OH)_4$. Thus, Puigdomenech and Bruno's log K for the reaction $Pu(OH)_4 + 4H^+ = Pu^{++++} + 4H_2O$ equals -0.8106, versus 0.7578 for GEMBOCHS. This would account for the lower Pu concentrations predicted by Bruno and Sellin.

Two solids, $Pu(OH)_4$(s) and $Pu(OH)_4$(am) appeared in the 0288 data base used by Bruno and Sellin. The data in Lemire and Tremaine and GEMBOCHS corresponds to that of $Pu(OH)_4$(s). It is possible that the data base from Puigdomenech and Bruno used by Bruno and Sellin retained the $Pu(OH)_4$(am) solid but deleted the $Pu(OH)_4$(s) solid. This is also consistent with the lower Pu concentrations predicted by Bruno and Sellin. Although seemingly contradictory, the $Pu(OH)_4$(am) solid was actually more stable than $Pu(OH)_4$(s). Equilibrium with respect to $Pu(OH)_4$(s) in version 0288 or $Pu(OH)_4$ in version comR16 rather than the $Pu(OH)_4$(am) used by Bruno and Sellin produces a very conservative value for Pu in solution.

The dominant aqueous species predicted by both codes are similar. The controversy over the validity of the existence of the aqueous species $Pu(OH)_5^-$ continues. Puigdomenech and Bruno do not include this species in their data base. However, for comparison and to evaluate its impact, its presence was considered in a separate set of calculations, and shown in Tables 1 and 2 in parentheses. The differences in Pu concentrations with and without $Pu(OH)_5^-$ in the GEMBOCHS data base are not large. For example, Pu concentrations in the oxidizing fresh Finnsjöwater (Table 2) equal 2.1e-7 molal with $Pu(OH)_5^-$ in the data base, and 1.6e-7 molal without $Pu(OH)_5^-$.

Neptunium

Thermodynamic data for Np

Bruno and Sellin (1992) state they use data from GEMBOCHS version 0288 for Np, but they apparently deleted the aqueous species $Np(OH)_5^-$. The GEMBOCHS version comR16 still contains this species. There appears to be a discrepancy as to the source of the Np data used by Bruno and Sellin. Bruno and Sellin (1992) state that the data sources in their EQ3/6 data base version 0288 are Lemire and Tremaine's (1980) article concerning U and Pu data, and Lemire (1988). In contrast, GEMBOCHS uses data from Lemire (1984).

Comparison of modeling results

The computed concentrations of Np in the reducing saline water differ by an order of magnitude. Under these conditions, and assuming that the species $Np(OH)_5^-$ does not exist, the solubility-controlling reaction is $Np(OH)_4(s) = Np(OH)_4^o$. Given this reaction, EQ3 and GEMBOCHS predict a concentration of 1.6e-9 molal, whereas Bruno and Sellin calculate 2e-8 molal. The log K for this reaction has not changed from the GEMBOCHS data bases dating back to the 0288 version to the present, so it is not clear why the LLNL and SKB concentrations differ. However, this may be a typographical error in Bruno and Sellin's Table 3. In the reducing fresh waters, Bruno and Sellin calculate 2e-9 molal, as does GEMBOCHS. It appears, therefore, that the molality of total Np in the reducing saline waters should equal 2e-9.

Computed concentrations of Np in both fresh and saline oxidizing waters are higher using GEMBOCHS than Bruno and Sellin's values, even though the Np speciation seems to be similar. The difference is barely significant in the fresh waters, but equals a factor of 20 in the saline waters.

Americium

Thermodynamic data for Am

Bruno and Sellin (1992) use the data from GEMBOCHS version 0288 for Am. Equilibrium constants for the hydrolysis and complexation reactions involving $AmCO_3^+$, $Am(CO_3)_2^-$, $AmOHCO_3(s)$ and other species in the 0288 data base used by Bruno and Sellin are very similar to those in GEMBOCHS version data0R16 used in this study.

Comparison of modeling results

GEMBOCHS calculates total Am concentrations about 2-3 times larger than those of Bruno and Sellin. A minor portion of this discrepancy might arise in the fresh waters because Bruno and Sellin's calculations do not appear to include provision for the Am complex $Am(CO_3)_2^-$. The increased carbonate content of the fresh waters favor formation of this complex. However, it comprises less than 10 mol% of the total Am in solution in the fresh waters, and it does not form in the saline waters, so it alone can not account for the discrepancies. Both sets of calculations utilized the EQ3/6 codes, so it is not believed that the differences could be attributed to differences in calculated activity coefficients.

Bruno and Sellin state that aqueous speciation is not affected by the salinity of the water. However, calculations with the GEMBOCHS data base suggest that in the fresh water, 100% of the Am is complexed by carbonate, whereas only about 50% is complexed by carbonate in the saline waters.

Technetium

Thermodynamic data for Tc

Despite the fact that Bruno and Sellin made some changes to the Tc data base, calculated Tc concentrations from the two data bases yield generally similar results. It is expected that under reducing conditions in both fresh and saline waters $TcO_2 \cdot 2H_2O(am)$ will be the solubility-limiting phase, rather than a number of phases more thermodynamically stable but not expected to form owing to kinetic considerations. Whereas Bruno and Sellin list TcO_2 as the solubility-limiting phase, GEMBOCHS predicts $TcO_2 \cdot 2H_2O(am)$. Bruno and Sellin may have omitted the $H_2O(am)$ suffix. Only $TcO_2 \cdot 2H_2O(am)$ exists in the 0288 data base used by Bruno and Sellin. In fact, the appropriate number of waters of hydration is problematic. It is expected that the numbers of water of hydration in $TcO_2 \cdot 2H_2O(am)$ will change to about 1.6 in the new Nuclear Energy Agency data base for Tc being compiled by J. Rard (Lawrence Livermore National Laboratory, pers. comm.).

Predicted concentrations in solution under reducing conditions in both fresh and saline waters using GEMBOCHS are about 1.7 times higher than those predicted by Bruno and Sellin. Under oxidizing conditions, both data bases predicted dominance of the species TcO_4^- in solution. All Tc-bearing solids were so soluble that no solubility-limiting phase could be identified in the models.

Conclusions

Differences have been identified between EQ3 predictions of solubility-limited radionuclide concentrations and aqueous speciation using the 0288 data base and other sources cited by Bruno and Sellin (1992) with those using GEMBOCHS version comR16. Predicted concentrations generally agree within a factor of 2 to 3, except when obvious differences in choices of thermodynamic data exist.

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TABLE 1. Comparison of solubility-limited concentrations at 25°C in the saline Finnsjöwater for U, Pu, Am, Np, and Tc calculated using EQ3/6 and the SKB and LLNL (GEMBOCHS version comR16) data bases.

Saline	Oxidizing (Eh = 650 mV)				Reducing (Eh = -200 mV)			
	Concentration limiting phase/ Concentration (molality)		Dominant aqueous species (%)		Concentration limiting phase/ Concentration (molality)		Dominant aqueous species (%)	
	SKB91	LLNL	SKB91	LLNL	SKB91	LLNL	SKB91	LLNL
U	schoepite $3x10^{-6}$	schoepite[a] $3.8x10^{-5}$	$UO_2(CO_3)_2^{2-}$ $UO_2(CO_3)_3^{4-}$	$UO_2(CO_3)_2^{2-}$ - 16 $UO_2(CO_3)_3^{4-}$ - 8 $(UO_2)_2CO_3(OH)_3^{1-}$ - 65 $UO_2(OH)_2^{0}$ - 8	UO_2(fuel) $2x10^{-7}$	UO_2(fuel)[b] $3.2x10^{-7}$	$U(OH)_4^o$	$U(OH)_4^o$ - 100
Pu	$Pu(OH)_4$ $3x10^{-8}$	$Pu(OH)_4$[c] $2.1x10^{-7}$ $(2.9x10^{-7})$[d]	PuO_2^+	PuO_2^+ - 99 (PuO_2^+ - 71 $Pu(OH)_5^-$ - 28)[d]	$Pu(OH)_4$ $5x10^{-8}$	$Pu(OH)_4$[c] $3.4x10^{-6}$ $(3.5x10^{-6})$[d]	Pu^{3+} $PuSO_4^+$	Pu^{3+} - 90; $PuSO_4^+$ - 7 (Pu^{3+} - 88: $PuSO_4^+$ - 6 $Pu(OH)_5^-$ - 2)[d]
Np	$NaNpO_2CO_3$ $4x10^{-4}$	$NaNpO_2CO_3 \cdot 3.5H_2O$[e] $7.9x10^{-3}$	NpO_2^+ NpO_2Cl	NpO_2^+ - 96 NpO_2Cl - 3	$Np(OH)_4$ $2x10^{-8}$	$Np(OH)_4$[f] $1.6x10^{-9}$ $(4.4x10^{-7})$[g]	$Np(OH)_4^o$	$Np(OH)_4^o$ - 100 $(Np(OH)_5^-$ - 100)[g]
Am	$AmOHCO_3$ $6x10^{-8}$	$AmOHCO_3$ $1.4x10^{-7}$	$AmCO_3^+$ Am^{3+}	$AmCO_3^+$ - 48 Am^{3+} - 26 $AmOH^{++}$ - 13	$AmOHCO_3$ $5x10^{-8}$	$AmOHCO_3$ $1.3x10^{-7}$	$AmCO_3^+$ Am^{3+}	$AmCO_3^+$ - 50 Am^{3+} - 25 $AmOH^{++}$ - 13
Tc	none high	none high	TcO_4^-	TcO_4^-	TcO_2 $2x10^{-8}$	$TcO_2 \cdot 2H_2O$-(am)[h] $3.3x10^{-8}$	$[TcO(OH)_2]_2$	$TcO(OH)_2^o$ - 84 $[TcO(OH)_2]_2^o$ - 16

a various U-bearing minerals are superaturated
b various U-bearing minerals are superaturated; thermodynamic data for UO_2(fuel) taken from Puigdomenech and Bruno, 1988, SKB Tech. Report 88-21
c PuO_2 is supersaturated
d if $Pu(OH)_5^-$ is considered
e NpO_2 and NpO_2OH(am) are superaturated
f NpO_2 is supersaturated
g if $Np(OH)_5^-$ is considered
h various Tc-bearing minerals are supersaturated

TABLE 2. Comparison of solubility-limited concentrations at 25°C in the fresh Finnsjöwater for U, Pu, Am, Np, and Tc calculated using EQ3/6 and the SKB and LLNL (GEMBOCHS version comR16) data bases.

Fresh	Oxidizing (Eh = 650 mV)				Reducing (Eh = -200 mV)			
	Concentration limiting phase/ Concentration (molality)		Dominant aqueous species (%)		Concentration limiting phase/ Concentration (molality)		Dominant aqueous species (%)	
	SKB91	LLNL	SKB91	LLNL	SKB91	LLNL	SKB91	LLNL
U	schoepite $3x10^{-4}$	schoepite[a] $2.6x10^{-4}$	$UO_2(CO_3)_2^{2-}$ $UO_2(CO_3)_3^{4-}$	$UO_2(CO_3)_2^{2-}$ - 40 $(UO_2)_2CO_3(OH)_3^{1-}$ - 53 $UO_2(CO_3)_3^{4-}$ - 4	UO_2(fuel) $2x10^{-7}$	UO_2(fuel)[b] $3.2x10^{-7}$	$U(OH)_4^o$	$U(OH)_4^o$ - 100
Pu	$Pu(OH)_4$ $3x10^{-9}$	$Pu(OH)_4$[c] $1.6x10^{-7}$ $(2.1x10^{-7})$[d]	PuO_2^+	PuO_2^+ - 98 $Pu(OH)_4^o$ - 1 (PuO_2^+ - 74 $Pu(OH)_5^-$ - 25)[d]	$Pu(OH)_4$ $2x10^{-8}$	$Pu(OH)_4$[c] $8.7x10^{-7}$ $(9.2x10^{-7})$[d]	Pu^{3+} $PuSO_4^+$	Pu^{3+} - 88; $PuSO_4^+$ - 7 $PuOH^{2+}$ - 5 (Pu^{3+} - 83; $PuSO_4^+$ - 7 $Pu(OH)_5^-$ - 6 $PuOH^{++}$ - 5)[d]
Np	NpO_2OH $2x10^{-3}$	NpO_2OH[e] $2.6x10^{-3}$	NpO_2^+ $NpO_2CO_3^-$	NpO_2^+ - 96 $NpO_2CO_3^-$ - 3	$Np(OH)_4$ $2x10^{-9}$	$Np(OH)_4$ $1.6x10^{-9}$ $(2.8x10^{-7})$[f]	$Np(OH)_4^o$	$Np(OH)_4^o$ - 100 ($Np(OH)_5^-$ - 100)[f]
Am	$AmOHCO_3$ $2x10^{-8}$	$AmOHCO_3$ $7.1x10^{-8}$	$AmCO_3^+$ Am^{3+}	$AmCO_3^+$ - 88 $Am(CO_3)_2^-$ - 8	$AmOHCO_3$ $2x10^{-8}$	$AmOHCO_3$ $7.1x10^{-8}$	$AmCO3^+$ Am^{3+}	$AmCO_3^+$ - 88 $Am(CO_3)_2^-$ - 9
Tc	none high	none high	TcO_4^-	TcO_4^-	TcO_2 $2x10^{-8}$	$TcO_2 \cdot 2H_2O$[g] (am) $3.3x10^{-8}$	$[TcO(OH)_2]_2^0$	$TcO(OH)_2^o$ - 84 $[TcO(OH)_2]_2^o$ - 16

a various U-bearing minerals are superaturated
b more stable phases not considered; thermodynamic data for UO_2(fuel) taken from Puigdomenech and Bruno, 1988, SKB Tech. Report 88-21
c PuO_2 is supersaturated
d if $Pu(OH)_5^-$ is considered
e NpO_2 is supersaturated
f if $Np(OH)_5^-$ is considered
g various Tc-bearing minerals are supersaturated

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